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MINERALOGY AND MINERAL CHEMISTRY OF VARISCAN GRANITOIDS FROM HIGHIŞ MTS. (APUSENI MTS., ROMANIA)

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ABSTRACT

The Apuseni Mts. is built up by nappe systems of continental crust origin (Apusenides), and oceanic crust related nappe systems (Transilvanides). The Apusenides were formed during the pre-Gosau tectogenesis and are built up by the autochthonous Bihor Unit and two overthrust units: the Codru and Biharia Nappe Systems, which contain granitoid intrusions of Variscan age. These major Alpine tectonic units are represented in the Highiş Mts. by the Finiş Nappe (Codru Nappe System) and the Highiş-Muncel Nappe (Biharia Nappe System). The granitoid rocks of the two nappe systems differ significantly: the intrusions of the Codru Nappe System are syenogranites of calc-alkali character with biotite of high Mg and low Al^{VI} content, accompanied by primary and secondary muscovites, while those of the Biharia Nappe System are syenogranites of subalkali character, with biotites of low Mg content, and primary muscovites.

Key words: mineral chemistry, Variscan granitoids, Codru Nappe System, Biharia Nappe System, Highiş Mts., Apuseni Mts., Romania

INTRODUCTION

A major proportion of the Pre-Neogene basement of the Apuseni Mts. (Romania) and the Pannonian Basin (Hungary) is built up by the Tisia Composite Terrane Alpine Megatectonic Unit. The crystalline mass of the Tisia Composite Terrane is characterised by granitoid ranges and anticline wings of middle and high grade metamorphites (Pál Molnár et al., 2001, 2002). The largest basement exposure within the Tisia Composite Terrane is represented by the Apuseni Mts. The Apuseni Mts. are partially built up by two Alpine overthrust units (Codru and Biharia Nappe Systems), carrying Variscan granitoid intrusions (Pană, 1998). These granitoids were mainly characterized by petrographical and geochronological studies (Giuşcă, 1979; Pană, 1998), their relation to the Pannonian Basin granites are less studied (Kovács et al., 2000).

The paper presents results of mineralogical and mineral chemistry studies performed on granitoids of the Codru and Biharia Nappe Systems, exposed in the Highiş Mts. (Fig. 1) The final aim of the research is to reveal correlations between the granitoids of the Apuseni Mts. and the variscan granitoids of the South Hungarian Basement.

GEOLOGICAL SETTING AND LOCATION

The Highiş Mts. are located on the W-SW part of the Apuseni Mts. Previous researches were made by Lóczy (1883); Rozlozsnik (1913); Paucă (1941); Giuşcă (1948, 1962, 1979); Giuşcă et al. (1964); Dimitrescu (1962, 1967, 1988); Savu (1965); Balintoni (1986, 1994); Tatu (1998); Pană (1998); Balintoni, Puşte (2002). Its crystalline basement is formed by the Tisia Composite Terrane, its main mass is made up by nappes of the Codru and Biharia Nappe Systems, both of which were formed during the pre-Gosau tectogenesis but with strikes of opposite direction (Fig. 2). The Codru Nappe System is in lower position, and Biharia is in upper position. Both nappe systems are positioned on the Biharia Unit (Săndulescu, 1984), and both contains granitoids of variscan age (Pană, 1998) (Fig. 2).

The granitoids of the Codru Nappe Sytem, which are located in the Highiş

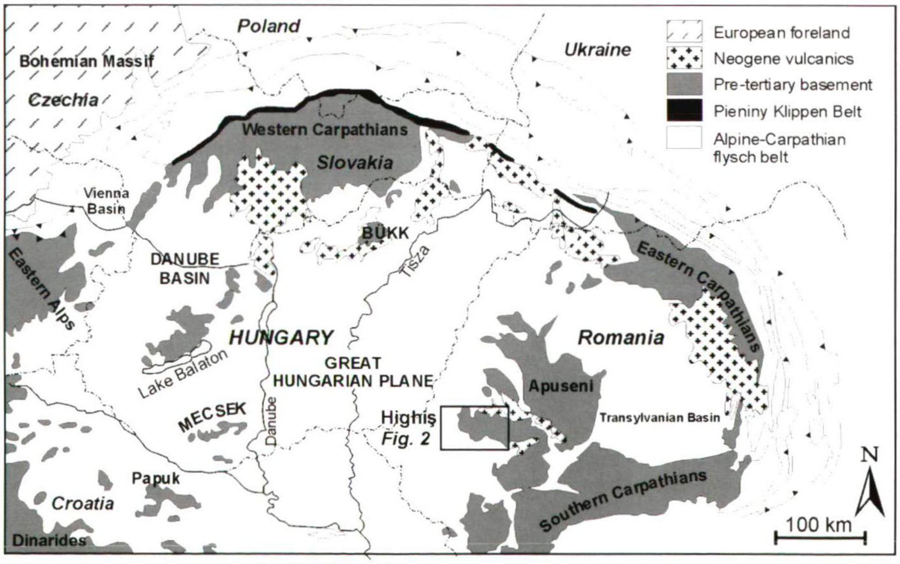

Fig. 1. The location of the Highiş Mts.

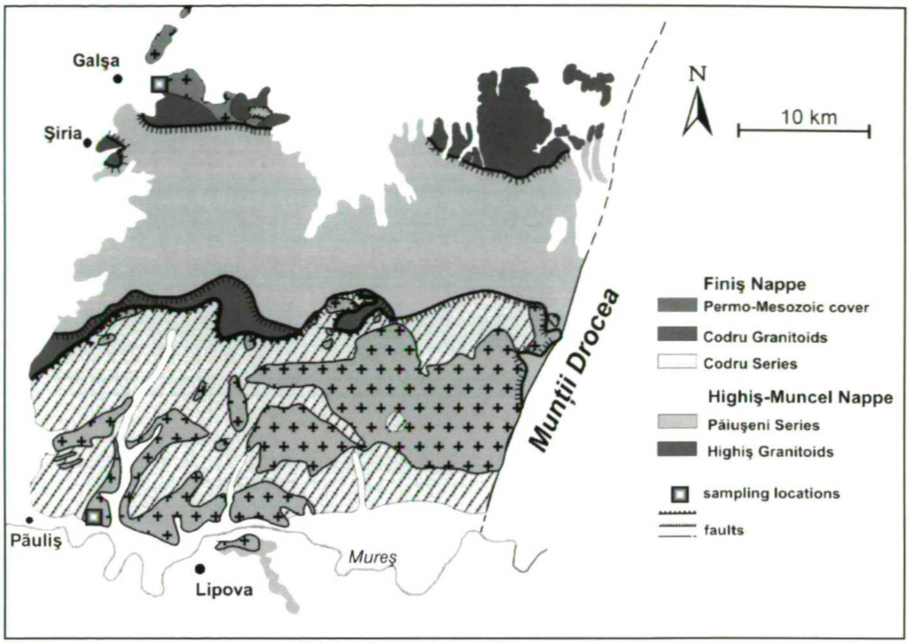

Fig. 2. The geologic map of the Highiş Mts.

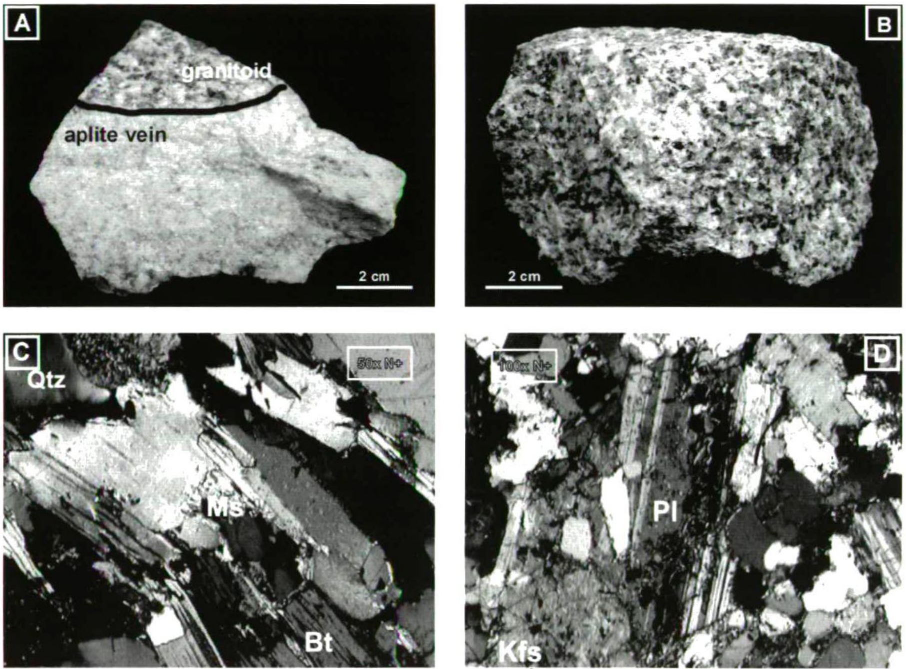

Fig 3. (A, B) Macroscopic view of the samples ÁGK-7272 (Galşa) and ÁGK-7269 (Păuliş); ***(C, D)*** Photomicrographs showing the mineral composition and texture of the samples ÁGK-7278 (Galşa) and ÁGK-7262 (Păuliş).

Mts. are positioned into the Upper Proterosoic Codru sequence as a part of the Finiş Alpine Nappe (Şiria granitoids). Şiria granitoids, located on the N and W part of the Highiş Mts., form a unified mass with a network of aplitic and pegmatitic veins, their contact zone is characterised by biotite-rich hornfels and paragneises of high biotite content. According to Pană (1998), their age is on the boundary of Carboniferous and Permian (Fig. 2). With the help of the K/Ar method Soroiu et al. (1969) determined a 221-226 Ma age from the biotites of the Şiria granitoides.

The granitoids of the Biharia Nappe System, which are located in the Highiş Mts., are positioned in the Biharia Lower Paleosoic sequence as a part of the Biharia Alpine Nappe. In their contact zones hornfelised metabasites and paragneises can be found (Highiş granitoids) (Fig.2). Highiş granitoids are Variscan, postcinematic granites, containing aplitic and pegmatitic veins (Giuşcă, 1979). Giuşcă et al. (1964) estimated a 350 Ma age from the Highiş Granitoid Complex with the help of K/Ar (WR) method. Nevertheless, Pană (1998) with the more reliable U/Pb method determined a 264-267 Ma age from zircon fractions, and he explained the formation of Highiş granitoids with a short lasting magmatism at the end of the early Permian.

SAMPLING AND ANALYTICAL METHODS

Samples are originating from the vicinity of settlement Galşa (Şiria granitoids - 32 rock samples) and Păuliş (Highiş granitoids - 28 rock samples) (Fig. 2). During the research 84 mineral chemical analyses were made at Department of Mineralogy and Petrology, University of Graz. Mesurments were performed at a 15 kV acceleration voltage and 10 nA current. Spectra were evaluated with software Oxford-Isis. Processing of raw data was made with softwares MinPet 2.0 and Minprog.

MINERALOGY AND MINERAL CHEMISTRY

On the basis of modal analyses, rock samples from Galşa are syenogranites with high mica content (10-12 tf%). The modal composition of Păuliş samples refers to syenogranites with a 1-3 tf% mica content (Le Maitre, 1989; not shown). Mineral chemical analyses were performed on feldspar, biotite and muscovite. In all 84 microprobe measurements were made: 36 on feldspars, 10 on biotites and 20 on muscovites (Table 1, 2 and 3). Samples coming from Galşa (Fig. 3) have a darker, greyish colour, their texture is phenocrystalline, porphyritic at some places. Syenogranites of aplitic texture are also represented. Rock forming minerals are equigranular. The colour of the Păuliş samples (Fig. 3) is pinkish, sometimes greyish, their texture is phenocrystalline, equigranular, medium-grained. Aplitic veins of greyish colour do also occur.

Table 1. Representative chemical composition of the studied feldspars.

Mineral	plag.	plag.	plag.	plag.	Kfs.	Kfs.	Kfs.	Kfs.	plag.	plag.	plag.	plag.	Kfs.	Kfs.	Kfs.	Kfs.
Sample	7271/11	7278/8a	7278/13	7278/18	7271/5	7271/8	7278/1	7278/7	7260/18	7262/5	7262/11	7264/3	7260/2	7260/17	7260/21	7264/2
	Galsa granitoids								**Paulis granitoids**							
Na_2O	10,70	10,42	9,33	8,60	0,52	0,62	0,65	1,23	10,44	10,38	9,54	10,98	0,55	0,70	0,31	0,30
MgO	0,42	0,00	0,09	0,00	0,35	0,14	0,06	0,10	0,05	0,29	0,22	0,19	0,22	0,23	0,00	0,16
Al_2O_3	20,22	19,79	21,57	22,25	18,21	17,82	18,15	18,05	19,90	19,44	19,21	19,97	18,00	17,60	17,64	17,92
SiO_2	67,26	68,96	66,00	63,60	63,43	63,55	63,75	63,82	70,49	69,60	69,89	69,87	63,99	63,70	65,17	64,74
K_2O	0,04	0,08	0,19	0,26	15,35	15,58	15,33	14,82	0,08	0,04	0,85	0,10	16,17	15,81	16,23	16,63
CaO	1,10	0,46	2,31	3,71	0,01	0,04	0,04	0,08	0,32	0,06	0,08	0,46	0,00	0,00	0,00	0,02
TiO_2	0,00	0,00	0,00	0,09	0,01	0,00	0,20	0,05	0,02	0,01	0,00	0,00	0,02	0,07	0,00	0,01
MnO	0,00	0,00	0,00	0,00	0,00	0,00	0,05	0,00	0,00	0,00	0,00	0,02	0,02	0,06	0,01	0,11
FeO	0,00	0,08	0,02	0,15	0,03	0,03	0,03	0,06	0,13	0,09	0,02	0,06	0,05	0,01	0,10	0,00
∑ oxides	99,75	99,80	99,51	98,65	97,90	97,77	98,27	98,21	101,43	99,91	99,82	101,65	99,02	98,18	99,47	99,89
	cation numbers based on 8 oxygens								cation numbers based on 8 oxygens							
Na	0,91	0,88	0,80	0,74	0,05	0,06	0,06	0,11	0,87	0,87	0,81	0,92	0,05	0,06	0,03	0,03
Mg	0,03	0,00	0,01	0,00	0,02	0,01	0,00	0,01	0,00	0,02	0,01	0,01	0,02	0,02	0,00	0,01
Al	1,04	1,02	1,12	1,17	1,01	0,99	1,00	1,00	1,01	1,00	0,99	1,01	0,99	0,98	0,97	0,97
Si	2,95	3,01	2,90	2,84	2,99	3,00	2,99	2,99	3,02	3,02	3,05	3,01	2,99	3,00	3,04	3,01
K	0,00	0,00	0,01	0,01	0,92	0,94	0,93	0,89	0,00	0,00	0,00	0,01	0,96	0,95	0,96	0,98
Ca	0,05	0,02	0,10	0,18	0,00	0,00	0,00	0,00	0,01	0,00	0,00	0,02	0,00	0,00	0,00	0,00
Ti	0,00	0,00	0,00	0,00	0,00	0,00	0,02	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00
Mn	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00
Fe	0,00	0,00	0,00	0,01	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00	0,00
O	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00	8,00
∑ cations	4,98	4,93	4,94	4,95	4,99	5,00	5,00	5,00	4,91	4,91	4,86	4,98	5,01	5,01	5,00	5,00
Or	0,23	0,49	1,09	1,52	95,15	94,25	93,90	88,44	0,43	0,18	5,51	0,52	95,09	93,70	97,18	97,30
Ab	94,42	97,20	87,00	79,57	4,80	5,60	5,95	11,16	97,92	99,51	94,11	97,23	4,91	6,30	2,82	2,66
An	5,35	2,31	11,91	18,91	0,05	0,15	0,15	0,40	1,65	0,31	0,37	2,25	0,00	0,00	0,00	0,04

Quartz: xenomorphic, 2-4 mm grains. As a result of deformation it is always undulating and frequently recrystallized, which lead to the decrease of grain size and the development of subgrains.

The representative chemical composition of feldspars can be studied in Table 1.

Orthoclase: hypidiomorphic, tabular habit, mean grain size is 4-5 mm, though in case of Galşa orthoclase crystals some 7-8 mm large megacrystals also occur. Twinning also appears (Carlsbad twins), twins occasionally have a perthitic structure. Microcline: hypidiomorphic, rarely xenomorphic, 3-4 mm grains, tabular habit.

The analysed potassium feldspars of Galşa syenogranites have a $Or_{87,28-95,39}Ab_{4,61-12,32}An_{0-0,40}$ composition. The Păuliş potassium feldspars represent $Or_{93,70-97,77}Ab_{2,23-6,30}An_0$ (Table 1; Fig. 4).

Plagioclase feldspars: hypidiomorphic, tabular, often zoned, mean grain size is 3-5 mm. Albite twins are common. Plagioclase feldspars of Galşa are albite-oligoclases ($An_{2,22-18,91}$). When examining the zoned plagioclase crystals of Galşa, an increased

Table 2. Representative chemical composition of the studied biotites.

Mineral	bio.	bio.	bio.	bio.	bio.	bio.	bio.	bio.	phl.	phl.	phl.	phl.
Sample	7271/9	7278/3	7278/4	7278/5	7260/ 10	7260/ 19	7264/ 12	7264/ 13	7262/2	7262/3	7262/6	7262/7
	Galşa granitoids				**Păuliş granitoids**				**Păuliş granitoids**			
Na_2O	0,07	0,04	0,04	0,09	0,05	0,05	0,15	0,08	0,05	0,07	0,17	0,05
MgO	8,81	8,68	8,98	9,16	5,43	5,27	5,33	5,39	14,30	13,62	14,67	15,26
Al_2O_3	15,35	16,23	15,89	16,10	13,42	14,12	13,52	14,10	13,38	13,56	13,79	13,95
SiO_2	35,64	34,82	34,93	35,28	34,88	35,87	34,95	34,11	36,84	36,08	40,76	40,51
K_2O	8,83	9,62	9,55	9,39	9,29	9,58	9,10	9,66	8,15	8,16	7,35	8,54
CaO	0,21	0,03	0,17	0,06	0,02	0,06	0,01	0,04	0,12	0,12	0,30	0,21
TiO_2	2,74	3,12	3,17	2,96	1,51	1,52	1,35	1,66	1,04	1,04	0,97	0,99
MnO	0,54	0,48	0,39	0,46	0,40	0,46	0,55	0,22	0,08	0,15	0,08	0,07
FeO	19,59	20,84	20,39	20,13	26,49	27,28	26,89	27,08	11,20	11,49	12,02	12,06
∑ oxides	91,79	93,84	93,52	93,62	91,49	94,22	91,85	92,34	85,16	84,28	90,10	91,65
	Cation numbers based on 22 oxygens											
Na	0,02	0,01	0,01	0,03	0,01	0,02	0,05	0,26	0,01	0,02	0,05	0,01
Mg	2,09	2,03	2,11	2,14	1,34	1,27	1,32	1,33	3,47	3,35	3,32	3,43
Al	2,88	3,01	2,95	2,97	2,62	2,69	2,64	2,75	2,57	2,64	2,47	2,48
Si	5,67	5,47	5,50	5,52	5,79	5,79	5,79	5,65	6,00	5,96	6,20	6,11
K	1,79	1,93	1,92	1,87	1,96	1,97	1,92	2,04	1,69	1,72	1,42	1,64
Ca	0,04	0,01	0,03	0,01	0,00	0,01	0,00	0,01	0,02	0,02	0,05	0,03
Ti	0,33	0,37	0,38	0,35	0,19	0,18	0,17	0,21	0,13	0,13	0,11	0,11
Mn	0,07	0,06	0,05	0,06	0,06	0,06	0,08	0,03	0,01	0,02	0,01	0,01
Fe	2,61	2,74	2,69	2,63	3,67	3,68	3,73	3,75	1,52	1,59	1,53	1,52
O	22,00	22,00	22,00	22,00	22,00	22,00	22,00	22,00	22,00	22,00	22,00	22,00
∑ cations	15,50	15,63	15,64	15,58	15,64	15,67	15,70	38,03	15,42	15,45	15,16	15,34
mg	44,47	42,56	43,96	44,86	26,75	25,66	26,14	26,18	69,54	67,81	68,45	69,29
Al^{IV}	2,34	2,53	2,53	2,47	2,20	2,21	2,21	2,35	2,00	2,04	1,79	1,89
Al^{VI}	0,54	0,48	0,44	0,50	0,43	0,48	0,44	0,40	0,57	0,60	0,68	0,60

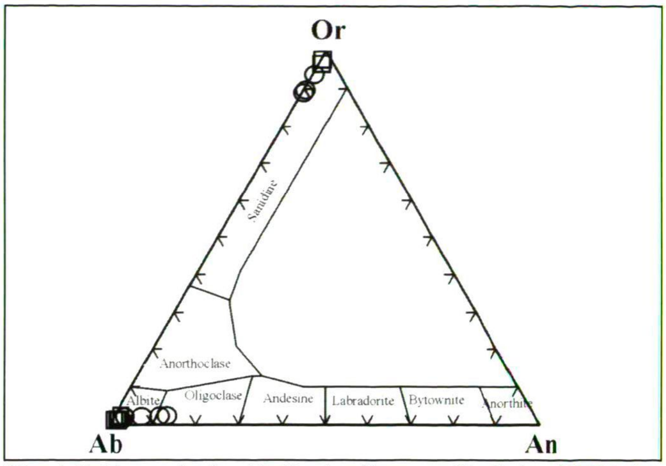

Fig. 4. Feldspars in the Ab-Or-An diagram. ○- Galşa; □-Păuliş

anorthite content can be detected in their centre compared to the margins: (ÁGK 7278/16-18: $An_{10,68-18,91}$, ÁGK7278/21b-a:$An_{13,75-16,06}$) (Table 1.). The Păuliş plagioclase feldspars are albites ($An_{0,30-1,65}$). The distribution of feldspars according to Or-Ab-An can be seen on Fig. 4.

BIOTITE GROUP

The representative chemical compositions of the minerals of the biotite group are presented in Table 2. Hypidiomorphic tabular or xenomorphic grains are characteristic, mean grain sizes are 2-5 and 1-3 mm in terms of the Galşa and Păuliş samples, respectively. Their pleochroism is light brown – dark green. If intergrown with muscovite they often contain opaque minerals, apatite and zircon. Along microtectonical deformations they have got a slight orientation. Biotites frequently group into nests. Based on Foster (1960), those samples were considered biotites where the sum of cations in X position and in Y position was between 1.60 – 2.20 (mean: 1.91) and between 5.30 – 5.28 (mean: 5.15), respectively. The average TiO_2 content is 2.91% in Galşa biotites and 0.90% in Păuliş biotites (Table 2). Galşa biotites have an ordinary Mg content, the value of the magnesium number (mg#) varies between 42.56 – 44.86 (mean mg#=44); (mg#=[Mg/(Mg+Fe)]x100, $Fe=Fe^{2+}+Fe^{3+}$). The Mg content of Păuliş biotites is low (mg#=25,15-27; mean mg#=26), and they are replaced by phlogopites along fractures, mg#= 67,81-69,54 (mean mg#=68,77) (Table 2., Fig 5B). Based on their Al^{IV} *vs.* Fe /(Fe+Mg) composition, the Păuliş and Galşa biotites form three well defined classes (Fig. 5A).

The Al^{VI} and Mg content of biotites provide information on the petrogenetics of granitoids. During magma fractionation the Fe and Al^{VI} content of the rock increases, while the Mg content decreases (Hecht, 1994). Biotites of Galşa granitoids are characterised by high Mg and low Al^{VI} content, which refers to a slightly fractioned magma, formed in the early phase of magma evolution (Fig. 5E). The Mg content of Păuliş biotites is low, which suggests a fractioned magma (Hecht, 1994) (Fig. 5E). The high Mg content of Păuliş phlogopites signs postgenetic transformations, which is also supported by the fact that samples containing phlogopite occur only along fractures, they are often weathered, and have a high muscovite content (in this case textural orientation is also apparent).

Table 3. Representative chemical composition of the studied muscovites.

Mineral	mus.	mus.	mus.	mus.	mus.	mus.	mus.	mus.
Sample	7271/7	7271/10	7278/2	7278/6	7260/15	7262/8	7262/9	7264/4
	Galşa granitoids				Păuliş granitoids			
Na_2O	0,55	0,34	0,36	0,25	0,83	0,20	0,09	0,14
MgO	0,74	0,74	0,82	1,05	2,10	3,35	3,35	1,29
Al_2O_3	33,85	31,35	31,55	30,24	25,61	27,41	28,62	28,53
SiO_2	46,46	45,45	46,83	46,16	47,13	50,52	50,29	48,70
K_2O	9,56	9,06	9,23	8,36	10,17	8,68	9,38	9,37
CaO	0,00	0,00	0,02	0,05	0,00	0,07	0,00	0,05
TiO_2	0,06	0,90	1,53	1,83	0,40	0,28	0,33	0,34
MnO	0,02	0,04	0,07	0,02	0,00	0,00	0,08	0,05
FeO	3,23	3,98	4,13	4,63	6,89	2,73	2,94	6,70
∑ oxides	94,48	91,87	94,54	92,58	93,13	93,24	95,09	95,17
	Cation numbers based on 22 oxygens							
Na	0,14	0,09	0,09	0,07	0,23	0,05	0,02	0,04
Mg	0,15	0,15	0,17	0,22	0,44	0,67	0,66	0,26
Al	5,37	5,14	5,03	4,91	4,25	4,36	4,50	4,56
Si	6,26	6,32	6,33	6,36	6,64	6,82	6,70	6,61
K	1,64	1,61	1,59	1,47	1,83	1,49	1,59	1,62
Ca	0,00	0,00	0,00	0,01	0,00	0,01	0,00	0,01
Ti	0,01	0,09	0,16	0,19	0,04	0,03	0,03	0,03
Mn	0,00	0,01	0,01	0,00	0,00	0,00	0,01	0,01
Fe	0,36	0,46	0,47	0,53	0,81	0,31	0,33	0,76
O	22,00	22,00	22,00	22,00	22,00	22,00	22,00	22,00
∑ cations	13,93	13,87	13,85	13,76	14,24	13,74	13,84	13,90

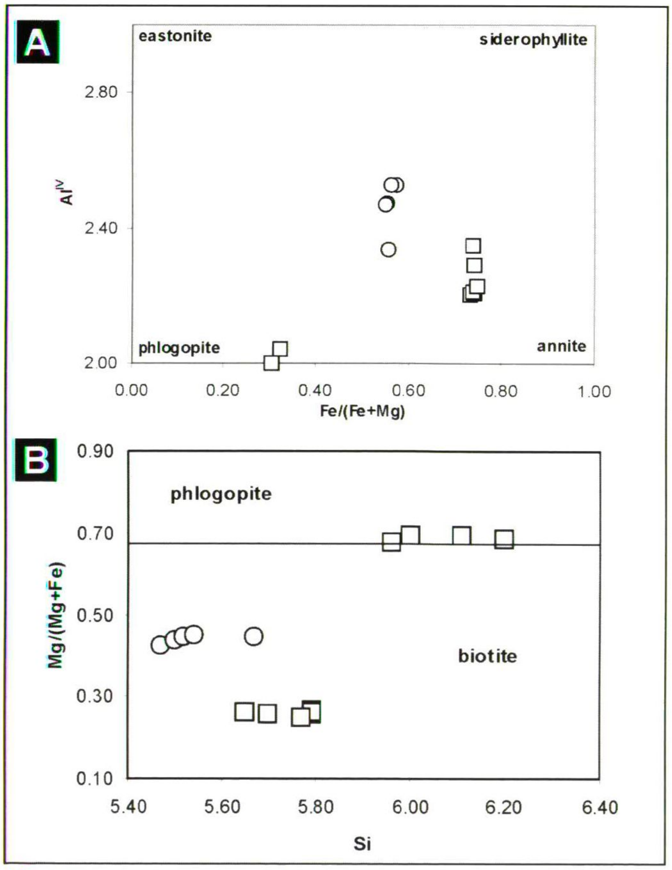

Fig. 5. (A) Compositional classification of the biotites; *(B)* Phlogopite/biotite discrimination diagram after Ferré, Leake (2001). ○- Galşa; □-Păuliş

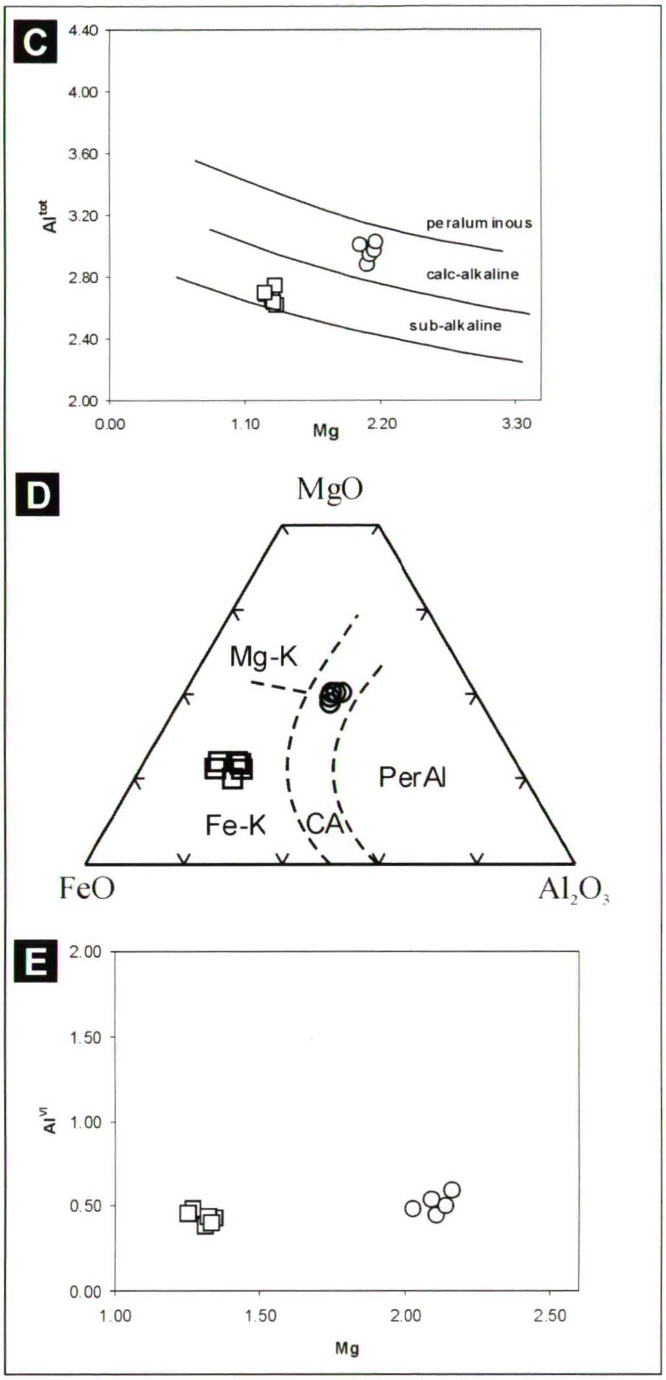

Fig. 5. (C) Mg vs. Al^{tot} diagram after Nachit et al (1985); ***(D)*** MgO vs. FeO vs. Al_2O_3 ternary diagram after Rossi and Chevremont (1987) and ***(E)*** Al^{VI} vs. Mg diagram after Hecht (1994). ○- Galşa; □-Păuliş

According to the Mg vs. Al_{tot} distribution of biotites (Nachit et al., 1985), the Galşa and Păuliş granitoids are of calc-alkali and subalkali character, respectively (Fig. 5C). The MgO-FeO*-Al_2O_3 distribution reinforces the calc-alkali character of Galşa syenogranites, while the Păuliş granites are originating from Fe-K (subalkali) magma (Rossi, Chevremont 1987) (Fig. 5D).

The representative chemical composition of muscovites can be studied in Table 3.

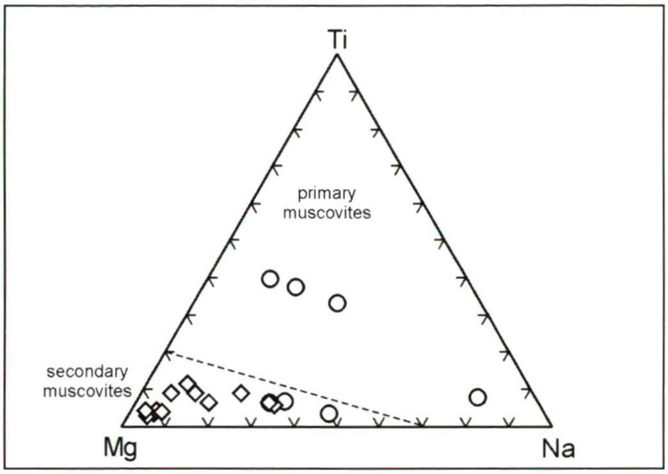

Fig. 6. Na-Mg-Ti muscovite classification after Miller et al. (1981). ○- Galşa; ◇-Păuliş

Muscovite: hypidiomorphic tabular and elongated lamellar, the mean grain size is 1–3 mm. It appears often along with biotite, and at some places it is oriented. Small sized muscovite grains are frequent in the fractures of the rock. Both group of samples contains ferrum-rich muscovites, the FeO content of Galşa samples varies between 3.23% and 4.36%, while that of the Păuliş samples is between 2.46% and 7.48%. The Păuliş muscovites have significant (1.29%–3.35%) magnesium content, however, in Galşa muscovites magnesium content is lower, than 1%. Based on the Na–Mg–Ti diagram (Fig. 6) (Miller et al., 1981), Galşa muscovites can be separated as muscovites of primary and secondary character, on the other hand Păuliş samples are uniform in representing only secondary character.

Accessory minerals are apatite, monazite and zircon.

Apatite crystals usually have an idiomorphic, partly hypidiomorphic shape, and often appear in biotite crystals.

Zircon crystals are idiomorphic, rarely hypidiomorphic, and represent two types of habit. One is squat, reddish-brown, yellowish-brown, the other is colourless, pinkish with an elongated columnar appearance. Opaque inclusions are quite frequent, numerous grains are zoned, which refers to several crystallisation phases.

Conclusions

Samples from both sites have got similar textural characteristics, i.e. they are holocrystalline, equigranular, medium-grained rocks (the only exceptions are aplites), textural orientation is unusual.

The main rock forming minerals of Galşa syenogranites are quartz, plagioclase feldspars (albite–oligoclase), orthoclase and microcline, biotite and muscovite. Accessory minerals are apatite and zircon. Based on their chemical composition, Galşa biotites are uniform.

The rock forming minerals of Păuliş granitoids are the following: quartz, orthoclase and microcline, plagioclase feldspar (albite) and biotite. Muscovite, apatite, monazite and zircon occur as accessory minerals. The Păuliş biotites can be divided into a Mg-poor group and secondary phlogopites. Galşa biotites are characterised by high Mg and low Al^{VI} content, which suggests an early phase of magma evolution.

Nevertheless, the Mg content of Păuliş biotites is low, which refers to a fractionated magma. A postgenetic phase can also be detected, which is signed by the high Mg content of biotites. According to the Mg vs. Al^{tot} distribution of biotites the Galşa syenogranites show a calc-alkali, the Păuliş syenogranites a subalkali character.

In terms of both granitoid groups muscovites have a significant Fe content (2.46%–7.48%). Galşa muscovites can be of primary or secondary character, however, Păuliş samples (based on the Na-Mg-Ti diagram) have a uniform secondary character.

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